PATENT APPLICATION
Navy Case No. **83,977**

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICATION FOR LETTERS PATENT

TO ALL WHOM IT MAY CONCERN:

BE IT KNOWN THAT **Ralph W. Bruce, Manfred Kahn, David Lewis III, Steven H. Gold and Arne W. Fliflet** who are citizens of the United States of America, and are residents of Arnold, MD, Myrtle Beach, SC, Alexandria, VA, New Carrollton, MD and Alexandria, VA, invented certain new and useful improvements in **"MICROWAVE ASSISTED REACTIVE BRAZING OF CERAMICS"** of which the following is a specification:

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MICROWAVE ASSISTED REACTIVE BRAZING OF CERAMIC MATERIALS

The present application claim the benefit of the priority filing date of provisional patent application No. 60/455,204 filed on 07 March, 2003, hereby incorporated, in its entirety, by reference.

FIELD OF THE INVENTION

This invention relates in general to the field of ceramics and in particular to the field of bonding high strength or high temperature ceramics.

BACKGROUND OF THE INVENTION

With the advent of new high tech materials, such as high temperature/high strength ceramics, the efficient joining of these materials is a major technical issue and very important to wide use of these materials. Many of these materials cannot be produced in large component sizes. Consequently, use of such materials in large scale devices requires joining of smaller components into assemblies of the required size.

Also it is difficult, if not impossible, to make high performance ceramic materials in complex shapes. The joining of simpler shapes on the other hand allows the preparation of more complicated shaped high performance ceramic parts, provided that the joints and surrounding regions exhibit, at all required operating conditions, the same strength and durability as the basic ceramic material.

Another difficulty with large scale ceramic assemblies or components can be their relative fragility compared to other metal or polymeric structures. Shipping of such large

assemblies from the site of manufacture to the site of use may well result in damage to the assembly and large replacement cost. One remedy for these problems is a robust capability for joining high temperature ceramics. This capability should include techniques for joining subassemblies in a production facility, but should also be capable of being used in a field setting, at the site of use.

One example of such a requirement is the production of large silicon carbide (SiC) heat exchanger assemblies for hot gas recuperation. This requires assembly of various tube segments, couplers, manifolds and other SiC and metal components into the large heat exchanger assembly. The joints must have sufficient strength to withstand the process pressures and handling during installation, and must also be gas tight. Another example is an accelerator application where there is a need to assemble a series of hot - pressed, high purity alumina tube segments, each about 3-4 cm in length, into a meter long dielectric-loaded accelerator assembly. In this case the joints need to have similar dielectric properties as the alumina, including very low loss and high breakdown voltage, and a pore free structure.

Another application is for joining ceramics plates to create complex structures such as conformal ceramic armor. If a low cost and efficient way to join ceramic plates such that the joint area shares similar physical and chemical characteristics with the base materials was available, complex, light weight multi-component assemblies could be assembled to conform to structures such as seats, engines, gearboxes or etc.

Hence, there exists a need to be able to join high performance ceramics, where the joint area shares the same or similar physical and chemical characteristics with the base materials.

SUMMARY OF THE INVENTION

A multi-component assembly featuring a structure with joints that share physical, chemical and electrical characteristics with the base materials and a process for creating a multi component structure by bonding base material parts using localized microwave energy and featuring the steps of:

Applying a thin coating of a joining material to each surface of the base materials to be joined; disposing each of the base materials such that the coated surfaces of the base materials being joined are in intimate contact in the desired alignment, and the pressure necessary to maintain the intimate contact and the desired alignment when joining the base materials and creating the desired component assembly; heating the joint area with a microwave beam applied to the surfaces of the base material being joined and focusing or diffusing the microwave beam to achieve localized heating of the joint area; heating the joint area to an initial joining temperature, wherein the joining material softens and fills physical discontinuities between the surfaces of the base materials being joined; rapidly heating the joint area to the reactive temperature of the joining material and the base materials; maintaining the joint area at the reactive temperature for a short interval to allow for the interdiffusion of the base and joining materials and formation of a homogenous joint region; rapidly cooling the joint area to a recrystalization temperature and maintaining the joint area at the recrystalization temperature for a predetermined

period; slowly cooling the joint area to room temperature, wherein the resulting component structure features joint region having similar physical, thermal and electrical characteristics as the base materials.

BRIEF DESCRIPTION OF THE DRAWINGS

FIGURE 1 shows the joint structure featuring the joining material applied to the surface of the base materials being joined.

FIGURE 2 shows the joint structure featuring the joining material and the base materials being joined during the initial heating phase of the joint area via a localized microwave beam.

FIGURE 3 shows the joint structure featuring the joining material and the base materials being joined during the reactive temperature phase.

FIGURE 4 shows the finished joint structure after recrystalization.

FIGURE 5 shows an example configuration of an apparatus for performing the microwave seamless joint bonding process at the heating stage.

DETAILED DESCRIPTION

Prior to a detailed description of the illustrations a brief overview of the process and of the resulting structure may be appropriate: Disclosed is a method for joining

material to efficiently create multi component assemblies in which the joints between the various components have virtually the same physical, chemical and electrical characteristics as the base materials. The process for creating such multi component structures uses a thin layer of a selected joining material applied to the juncture and localized microwave energy focused thereon to bond the component materials, and to diffuse away the joining material into the base material.

A thin coating of the joining material is applied to the surfaces of the base materials being joined. The base materials are disposed in relation to each other such that the two surfaces of the base materials being joined are in contact with each other and feature the desired alignment, and pressure necessary for maintaining the contact and the alignment required for the desired component assembly. The actual magnitude of the pressure applied during joining makes a minor contribution to the amount of materials diffusion that causes the joining during the heating process. Temperature and duration of the heating and cooling cycles are its primary determinants.

A microwave beam is focused on the joint area. The joint area is the area defined by the surfaces of the base material being joined. The microwave beam incident on the joint area is focused to achieve localized heating of the joint area. The surfaces beyond the joint area remain cool.

The joint area is heated to an initial joining temperature, wherein the joining material softens and fills physical discontinuities between the surfaces of the base materials being joined. The joint area is then rapidly heated to the reactive temperature of the joining material and the base materials. The joint area is maintained at the reactive

temperature for a short interval to allow for the reaction and interdiffusion of the base and joining materials and for the subsequent formation of a homogenous joint region.

Next the joint area is rapidly cooled to a recrystalization temperature and the joint area is maintained at the recystalization temperature for a predetermined period. This allows the joint region, now featuring the base materials regions interdiffused with each other and with the joining material, to form a homogeneous stable physical and thermal structure having uniform grain sizes. This process can be used to create very complex structures through sequential joining since the previous made joints are not significantly heated. Even if their temperature is increased, their properties would not be affected any differently than those of the base material, since the joining material by that time is expected to have diffused away.

With reference to the figures in which like reference numbers denote like elements, Figure 1 illustrates an example joint structure featuring the joining material applied to the surface of the base materials being joined. The process for bonding the base materials and creating the seamless joint structure employs localized microwave energy for heating. The microwave energy is focused on the joint area, causing the localized heating of the selected area only. The other surfaces of the base materials remain near the ambient temperature.

Figure 1 shows an example embodiment of the joint structure during the initial steps of the joining process. Base materials, 150, 170 are disposed to be in intimate contact with each other after a joining material 140 is applied to the joint area 160 of the base materials 150, 170 being joined.

Generally the base materials 150, 170 comprise high temperature ceramics which may be similar or dissimilar in chemical composition, shape and attributes. In a preferred embodiment these base materials are high temperature ceramics having a purity of greater than 99%. The base materials may be high temperature ceramics in the form of high purity oxide materials, however oxides are not required. In addition, the base material may be dissimilar materials.

As illustrated in Figure 1, a thin coating of a joining material 140 is applied to each surface of the base materials, 150 and 170 being joined. The coating is applied to the joint region 160, specifically to the joint surfaces 130 of the base materials to be joined.

The joining material is a frit or reactive braze selected such that at a predetermined temperature the constituents of the joining material will chemically react and interdiffuse with the base material. As a result of the frit or braze constituents diffusing away into the base materials, their concentration is reduced to undetectable levels, so that only the high temperature ceramic material remains, including in the joint area. The joining material is chosen to be chemically reactive with the base material at a temperature below that of the thermal degradation threshold of the base material.

In an example embodiment, wherein the base materials are high temperature oxides, the joining material would preferably be an oxide glass frit. The use of such joint material permits assembly with minimal bonding pressure, since the glass initially softens and flows during the bonding process. The use of an oxide glass frit also relaxes the requirements for fit and finish in joints - surface flatness and roughness, since the reactive glass frit can fill surface irregularities in the joint. With the use of localized heating, the

process can be used to create complex, multi surface structures through sequential heating, since previously made joints are homogenized by the diffusion process to have essentially the same composition and high temperature tolerance as the original base ceramic.

Referring again to Figure 1 and to Figure 5, each base material 150, 170 is disposed such that the surface of the base materials being joined 130 are in contact, with the desired alignment, and pressure necessary for joining the base materials and creating the desired component assembly. Figure 5 illustrates an example system as employed to create a multi component assembly according to the method described herein. Fixtures 580, 580' are employed to dispose each piece of base material 150, 170 such that the surface of the base materials being joined are in contact with the desired alignment, and pressure necessary for joining the base materials 150, 170 and creating the desired component assembly.

The fixtures 580, 580' may be low temperature-type fixtures and are used to position the respective pieces of the base materials 150, 170 with the desired alignment and pressure to create the desired joined structure.

Once the base materials 150, 170 have been coated at the surfaces to be joined and aligned, the next phase involves heating the joint area 160 with a microwave beam 220 applied to the surfaces of the base material being joined. The heating effect may be controlled by focusing or diffusing the microwave beam 220 to achieve localized heating of the joint area 160.

Figure 2 illustrates the joint structure featuring the thin layer of the joining material 140 and the base materials being joined 150, 170 during the initial heating phase of the joint area 160 via a localized microwave beam 220.

Microwave beam heating of materials offer a number of advantages in material processing and particularly in joining operations. One is much shorter processing times than in conventional furnaces. With focused millimeter-wave heating of materials, only the relatively small joint area is heated, minimizing effects of the thermal inertia of the system. This permits very rapid temperature changes. Typically in microwave and millimeter wave systems heating and cooling rates are only limited by two factors - thermal shock of the workpiece and the ability of the workpiece to dissipate heat to the environment during cooling.

Another major factor in the beam system employed is the ability to localize heating. This is clearly impossible in a conventional furnace or in a microwave cavity system. By empoying optics as in the present method, one can focus the beam down to a spot the size of a centimeter. The beam is focused, defocused or shaped by suitable reflective optics. The optics may even be equipped with motorized drives, if desired. This localization of the microwave beam has two important consequences. The first is that the heating can be confined to the joint area in an assembly being joined by this process, minimizing or eliminating heating of the remainder of an assembly. Confining the heating to the joint area eliminates thermal damage to thermally sensitive sub-elements of an assembly, while providing sufficiently high temperatures in the joint region to achieve a high strength, refractory joint in high temperature base materials.

Another related effect which may be used to achieve even greater localization of heating, is that with suitable dielectric properties, the incident millimeter wave beam can be guided down the joint itself, with essentially no energy deposited adjacent to the joint. This effect may be used to heat joints much smaller than the wavelength of the radiation used in current microwave generating "gyrotron" system.

Referring now to Figure 2 and with continued reference to figure 5, localized microwave beam heating is applied to the joint area 160 of the base materials 150, 170. The joint is heated to an initial joining temperature. As shown in figure 2, at the initial joining temperature the joining material 140 softens and fills physical discontinuities between the surfaces of the base materials 150, 170 being joined. In the example embodiment, the initial joining temperature is in the range of approximately 800 to 1200 degrees Centigrade depending on the materials selected.

Next the joint area is rapidly heated to the reactive temperature of the joining material and the base materials. The reactive temperature is defined as the temperature at which the joining material and the base materials chemically react. This temperature is generally greater than the initial joining temperature. The joint area is heated from the initial joining temperature to the reactive temperature at a rate of approximately 100 degrees Centigrade a minute. In the example embodiment, the reactive temperature is approximately 1400 to 1700 degrees Centigrade.

The joint is then maintained at the reactive temperature for a short interval to allow for the interdiffusion of the base and joining materials and formation of a homogenous joint region.

Figure 3 shows an example joint structure featuring the joining material and the base materials being chemically joined during the reactive temperature phase. As shown in the figure, base materials 150 and 170 are separated by a homogenous joint region 160 in which the constituents of the base materials and the joining material become interdiffused. Specifically base material 150 contains constituents represented by the letter "A" 151, while base material 170 contains constituents represented by the letter "C" 171. Base material 140 contains constituents represented by "B" 141. As shown in the figure constituents 151 diffuse into the join area 160 and the base material 170 while constituents 171 from base area 170 diffuse into the joint area 160 and into the base material 150. The constituents of the joining material 141 simultaneously diffuse into the base materials 150, 170, facilitating the formation of a homogenous joint. In a preferred embodiment the joint area is maintained at the reactive temperature for an interval of approximately 10 minutes or more.

After a completed joining operation, constituents "B" 141 are fully diffused into base materials 150 and 160. Their concentration in the joint region 160 is diluted to the extent that they have only a negligible and undiscernible effect on the properties of the joint. The joint then contains only homogenized constituents of the high performance base materials "A" 151 and "C" 171.

An additional benefit of use of localized heating by a directed microwave beam during the heating processes is that metal fixtures used to position and hold each base material do not generally reach a temperature above 100 degrees Centigrade. Generally, when other types of heating sources are used, heating effects cannot be contained to only the joint area, thus special heat resistant fixtures must be employed such as fixtures

materials 150, 170. Once the joined structure or component assembly is created, the process may be repeated as many times as necessary allowing subsequent component assemblies or base materials to be added to create the desired multi-component structure.

For purposes of example two or more thin pieces of high purity Alumina, such as WESGO 995, may be joined using the above method to form a complex compound structure. This alumina is of a very high purity, greater than 99.5%. The surfaces of the pieces of alumina to be joined are held together in intimate contact at a low pressure using low temperature fixtures such as those illustrated in figure 5. The joint surfaces are thinly coated with a glass frit. In this example embodiment, the frit material selected is a Calcia/Boria/Silica compound, such as the Fusion 588 glass frit. The frit comprises $CaO{\bullet}B_2O_3{\bullet}SiO_2$. This material was chosen because of it becomes chemically reactive with the base alumina at a temperature as low as 900 degrees C in combination with the frits softening at even lower temperatures.

A localized millimeter wave beam is focused on the joint area, and the joint area is initially heated to approximately 1200 degrees Centigrade. This is the initial joining temperature. At the initial joining temperature the frit material softens and fills physical discontinuities between the surfaces of the base alumina being joined.

Next the joint area is rapidly heated to the reactive temperature, which for purposes of this example, is approximately 1700 degrees Centigrade. The joint area is heated from the initial joining temperature to the reactive temperature at a rate of approximately 100 degrees Centigrade a minute. Once the joint area reaches the reactive temperature, the joint area's temperature is maintained at a constant 1700 degrees Centigrade for approximately 2 minutes. This provides ample time for the alumina and frit materials to interdiffuse.

For purposes of example two or more thin pieces of high purity Alumina, such as WESGO 995, may be joined using the above method to form a complex compound structure. This alumina is of a very high purity, greater than 99.5%. The surfaces of the pieces of alumina to be joined are held together in intimate contact at a low pressure using low temperature fixtures such as those illustrated in figure 5. The joint surfaces are thinly coated with a glass frit. In this example embodiment, the frit material selected is a Calcia/Boria/Silica compound, such as the Fusion 588 glass frit. The frit comprises $CaO{\bullet}B_2O_3{\bullet}SiO_2$. This material was chosen because of it becomes chemically reactive with the base alumina at a temperature as low as 900 degrees C in combination with the frits softening at even lower temperatures.

A localized millimeter wave beam is focused on the joint area, and the joint area is initially heated to approximately 1200 degrees Centigrade. This is the initial joining temperature. At the initial joining temperature the frit material softens and fills physical discontinuities between the surfaces of the base alumina being joined.

Next the joint area is rapidly heated to the reactive temperature, which for purposes of this example, is approximately 1700 degrees Centigrade. The joint area is heated from the initial joining temperature to the reactive temperature at a rate of approximately 100 degrees Centigrade a minute. Once the joint area reaches the reactive temperature, the joint area's temperature is maintained at a constant 1700 degrees Centigrade for approximately 2 minutes. This provides ample time for the alumina and frit materials to interdiffuse.

The joint area is then cooled to the annealing/recrystalization temperature. For a structure using the selected alumina and a $CaO{\bullet}B_2O_3{\bullet}SiO_2$ frit material this temperature is

approximately 900 degrees C. The joint area is maintained at this temperature for approximately 30 minutes.

The joint area is then allowed to cool to room temperature. The two pieces of Alumina are now joined creating an Alumina compound structure featuring a homogenous joint which shares physical and chemical characteristics very similar to the base alumina.

Once the alumina compound structure has been allowed to cool, complex multi-component assemblies may formed by joining the alumina compound structure with another base material or compound substructure using the above method.

Although this invention has been described in relation to the exemplary embodiment's thereof, it is well understood by those skilled in the art that other variations and modifications can be affected on the preferred embodiment without departing from scope and spirit of the invention as set fourth in the claims.